

## EPOXY RESIN COMPOSITION AND PREPREG AND PRINTED CIRCUIT BOARD USING THE SAME

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### Abstract

**PROBLEM TO BE SOLVED:** To obtain an epoxy resin composition capable of imparting good flame retardancy substantially without containing halogen atoms when forming an insulating layer by being supported to a sheetlike organic fibrous substrate.

**SOLUTION:** This epoxy resin composition comprises (A) an epoxy resin without containing a halogen, (B) a phenolic resin, (C) a thermosetting resin having dihydrobenzoxazine ring, (D) a phosphorus compound and (E) an inorganic filler as essential components. The insulating layer is formed by impregnating the epoxy resin composition into an aramid fibrous substrate (preferably polyparaphenylene terephthalamide fibrous substrate) and supporting thereto. The component (B) is preferably a phenolic novolak resin containing phosphorus atom or nitrogen atom in the molecular structure thereof.

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**DETAILED DESCRIPTION**

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[Detailed Description of the Invention]

[0001]

[The technical field to which invention belongs] this invention relates to the thermosetting resin constituent used as raw materials, such as the highly efficient molding material which is excellent in thermal resistance, fire retardancy, etc., a paint, a coating material, adhesives, a sealing agent, a prepreg for printed circuit boards, a metal tension laminate, FRP; and a carbon product, and its hardened material.

[0002]

[Description of the Prior Art] Thermosetting resin, such as phenol resin, melamine resin, an epoxy resin, an unsaturated polyester resin, and a bismaleimide resin, is used in many industrial fields with the thermal resistance and reliability based on the property of the thermosetting. However, phenol resin and melamine resin generate an volatile by-product at the time of hardening, an epoxy resin and an unsaturated polyester resin are inferior to thermal resistance, and the bismaleimide resin is very expensive -- etc. -- the respectively peculiar trouble existed and a compromise needed to be actually reached suitably about the trouble according to the use. Then, development of the new thermosetting resin which does not have these troubles has been furthered conventionally.

[0003] There is a dihydrobenzo oxazine compound as one of them. (JP,49-47387,A, U.S. Pat. No. 5152939 specification). Since hardening of this compound is a thing using the ring-opening-polymerization reaction of a dihydrobenzo oxazine ring, it heat-hardens without hardly being accompanied by generating of volatile matter.

[0004] Moreover, as the hardenability of these resins, and reactant research, there is reference of a report of Burke and others of J.Am.Chem.Soc.3424 (1965), Riess and others of Polym.Sci.Techol.27 (1985). Moreover, in case the phenol nature hydroxyl in a molecule is formed into dihydrobenzo oxazine, the dihydrobenzo oxazine compound which has improved hardenability is indicated by JP,7-188364,A by leaving without cyclizing a phenol nature hydroxyl at a specific rate.

[0005] Moreover, the dihydrobenzo oxazine compound which raised hardenability, without reducing a mechanical characteristic, and the thermosetting resin constituent containing novolak type phenol resin are indicated by JP,9-272786,A.

[0006]

[Problem(s) to be Solved by the Invention] As compared with the thermosetting resin by which the hardened material of the dihydrobenzo oxazine compound using the ring-opening-polymerization reaction indicated by JP,49-47387,A is known conventionally, thermal resistance is good and, moreover, it is high intensity. However, hardening by the ring-opening-polymerization reaction has the fault of having a long time compared with hardening by the hardening reaction of usual phenol resin, and the technical problem that the use on industry is limited in respect of productivity is also known.

[0007] Moreover, although it is checked that the hardenability of the benzo oxazine compound obtained is improvable when forming dihydrobenzo oxazine and it leaves a phenol nature hydroxyl at a certain fixed rate into a molecule as indicated by JP,7-188364,A, since ring opening reaction advances by heating at the time of composition of this compound, the technical problem that it is difficult to perform stable composition occurs.

[0008] Moreover, although the example of hardenability examination at the time of using a 1 functionality phenolic compound is indicated by the report of Burke, Riess and others, hardenability improvement is not enough and the fall of the thermal resistance by the fall of crosslinking density and a mechanical strength is accepted.

[0009] Although improvement of the hardenability of a dihydrobenzo oxazine compound was tried by combined use of novolak type phenol resin, in order to have applied to the molding material etc., the hardenability of JP,9-272786,A was still inadequate.

[0010] this invention aims at offering the constituent of the thermosetting resin which has the dihydrobenzo oxazine ring which raised a moldability and hardenability, without falling many properties, such as a fall of a mechanical characteristic, and its hardened material.

[0011]

[Means for Solving the Problem] That is, this invention is the thermosetting resin which has a (1) (A) dihydrobenzo oxazine ring, (B) phenol resin, and a thermosetting resin constituent which consists of a (C) Lynn system hardening accelerator.

(2) The thermosetting resin constituent of a phenol novolak resin, a phenol aralkyl resin, biphenyl denaturation phenol resin, xylene denaturation phenol resin, melamine denaturation phenol resin, and guanamine denaturation phenol resin given in (1) given phenol resin (B) is either at least.

(3) (1) whose thermosetting resin (A) which has a dihydrobenzo oxazine ring is thermosetting resin compounded from phenols, formaldehyde, and aromatic amines, or a thermosetting resin constituent given in (2).

(4) A thermosetting resin constituent given in (3) of the - diamino diphenyl-ether, 1, and 3-screw (4-amino phenoxy) benzene, and aniline, 4, and 4'-diamino diphenylmethane, 4, and 4'alpha, alpha'-screw (4-amino phenyl) -1 and 4-diisopropylbenzene whose aromatic amines used for the thermosetting resin (A) which has a dihydrobenzo oxazine ring are either at least.

(5) The hardened material which makes either come to harden the thermosetting resin constituent of a publication among (1) - (4) is offered.

[0012] Moreover, this invention offers the hardened material which comes to harden the thermosetting resin constituent of the above-mentioned this invention.

[0013] As a result of repeating examination wholeheartedly, by blending novolak type phenol resin with the thermosetting resin which has a dihydrobenzo oxazine ring as a curing agent, and blending the hardening accelerator of the Lynn system further, this invention persons harden without hardly being accompanied by generating of volatile matter, find out that a thermosetting resin constituent with many good properties, such as a mechanical property, is obtained, and came to complete this invention based on these knowledge.

[0014]

[Embodiments of the Invention] Although there is especially no limit, as for the thermosetting resin which has the dihydrobenzo oxazine ring of the (A) component used in this invention, what is compounded from phenols, aldehydes, and aromatic amines is desirable.

[0015] (A) As phenols used for the composition of thermosetting resin which has the dihydrobenzo oxazine ring of a component If it is the compound which has a phenolic hydroxyl group, although there will be especially no limit For example, a phenol, o-cresol, m-cresol, p-cresol, A xylenol, ethylphenol, p-t-butylphenol, a nonyl phenol, Monovalent alkylphenols, such as an octyl phenol, a resorcinol, A catechol, biphenol, bisphenol A, Bisphenol F Naphthols, such as polyhydric phenols, such as Bisphenol S and tetrafluoro bisphenol A, an alpha naphthol, a beta naphthol, and dihydroxy naphthalene Or phenol derivatives, such as a phenylphenol, an aminophenol, and a halogenation phenol, etc. are mentioned, and even if it uses these one sort independently, you may use combining two or more sorts.

[0016] (A) Although there will be especially no limit if it is the compound or aldehyde field which has an aldehyde group as aldehydes used for the composition of thermosetting resin which has the dihydrobenzo oxazine ring of a component, formaldehyde, formalin, a paraformaldehyde, etc. are mentioned, for example. The thing of 80% of the weight or more of formaldehyde concentration, especially the thing of a paraformaldehyde of 92% of the weight or more of formaldehyde concentration are desirable.

[0017] (A) As aromatic amines used for the composition of thermosetting resin which has the dihydrobenzo oxazine ring of a component If it is the amine derivative of an aromatic hydrocarbon, although there will be especially no limit For example, an aniline, a toluidine, an anisidine, m-phenylenediamine, A - diamino diphenylmethane, and p-phenylene diamine, 4, and 4'4, 4'-diamino diphenyl ether, 4 and 4' screw [-diaminodiphenyl-sulfone, 2, and 2-] [(4-amino phenoxy)phenyl] propane, Aromatic diamines, such as the 1, 3-screw (4-amino phenoxy) benzene, alpha, and alpha'-screw (4-amino phenyl) -1 and 4-diisopropylbenzene (common-name screw aniline-M), are mentioned, and even if it uses these one sort independently, you may use combining two or more

sorts. Especially, the 1 from fire-resistant viewpoint that - diamino diphenyl ether is desirable, 3-screw (4-amino phenoxy) benzene, and 4 from viewpoint of workability and the improvement in property, 4'-diamino diphenylmethane and 4, and 4'alpha, alpha'-screw (4-amino phenyl) -1, and 4-diisopropylbenzene are desirable.

[0018] When using a diamine, the thermosetting resin (A) which has two or more dihydrobenzo oxazine rings can be manufactured because the \*\*\*\* case for monoamines uses bisphenol A, F, and novolak type phenol resin for univalent phenols again.

[0019] As the synthetic method of thermosetting resin (A) of having a dihydrobenzo oxazine ring, although there is especially no limit, the method to which phenols, aldehydes, and aromatic amines are made to react in a solvent is mentioned. Especially, after making phenols and aldehydes suspend in a solvent, warm at 50-70 degrees C, add aromatic amines over 15 - 30 minutes, and carry out the temperature up of the reaction temperature to reflux temperature (about 80 degrees C) after that, it is made to react more preferably after emulsification for about 2 hours for 1 to 4 hours, and the method of removing a solvent and moisture under reduced pressure is desirable after a reaction end. When there is an inclination for a reaction to progress at less than 50 degrees C as aromatic amines and aldehydes do not dissolve, and for unreacted aromatic amines and aldehydes to remain in the system of reaction and the temperature at the time of aromatic-amines addition exceeds 70 degrees C, a reaction progresses partially and there is an inclination for a uniform resin to be hard to be obtained.

[0020] As a solvent used for a reaction, although there is especially no limit, lower alcohols, such as a methanol, ethanol, propanol, and a butanol, etc. are mentioned, and a methanol is desirable [ a limit ] from a viewpoint of the compatibility of a price and aldehydes especially.

[0021] (A) 150 degrees C or more of catalysts or curing agents are not preferably used for the thermosetting resin which has the dihydrobenzo oxazine ring of a component by heating at 170-220 degrees C -- also coming out -- it can be made to harden, without producing a by-product

[0022] Even if it uses one sort independently, you may use the thermosetting resin (A) which has a dihydrobenzo oxazine ring combining two or more sorts.

[0023] The phenol resin of the (B) component used in this invention Although there is especially no limit, for example A phenol, cresol, a resorcinol, A catechol, bisphenol A, Bisphenol F, a phenylphenol, Phenols, such as an aminophenol, and/or the alpha-naphthol, the beta-naphthol, Naphthols, such as dihydroxy naphthalene, and the compound which has aldehyde groups, such as formaldehyde, under an acid catalyst Condensation or the phenol novolak resin which is made to carry out copolycondensation and is obtained, The phenol aralkyl resin compounded from a naphthol novolak resin, phenols and/or naphthols, dimethoxy paraxylene, or a screw (methoxymethyl) biphenyl, A naphthol aralkyl resin, xylene denaturation phenol resin, naphthol denaturation phenol resin, Naphthalene denaturation phenol resin, biphenyl denaturation phenol resin, melamine denaturation phenol resin, guanamine denaturation phenol resin, etc. are mentioned, and even if it uses these independently, you may use combining two or more kinds. Especially, from a viewpoint of hypoviscosity-izing and a raise in restoration, a phenol aralkyl resin, xylene denaturation phenol resin, and biphenyl denaturation phenol resin are desirable, and melamine denaturation phenol resin and guanamine denaturation phenol resin are desirable from a fire-resistant viewpoint. When [ of a phenol aralkyl resin xylene denaturation phenol resin, biphenyl denaturation phenol resin, melamine denaturation phenol resin, and guanamine denaturation phenol resin ] using either at least, in order to demonstrate the performance, as for the loadings, it is desirable to consider as 60 % of the weight or more in all to the phenol resin whole quantity.

[0024] As a Lynn system hardening accelerator used by this invention Tributyl phosphine, a dibutyl phenyl phosphine, butyl diphenylphosphine, Ethyl diphenylphosphine, methyl diphenylphosphine, triphenyl phosphine, A tree p-tolyl phosphine, a tris (4-methylphenyl) phosphine, A tris (4-methoxyphenyl) phosphine, a tris (2, 6-dimethoxy phenyl) phosphine, In organic phosphines and these organic phosphines, such as diphenylphosphine and a phenyl phosphine, a maleic anhydride, The 1, 4-benzoquinone, 2, 5-torr quinone, 1, 4-naphthoquinone, 2, 3-dimethyl benzoquinone, 2, 6-dimethyl benzoquinone, 2, the 3-dimethoxy-5-methyl-1, 4-benzoquinone, Quinone compounds, such as 2, 3-dimethoxy-1, 4-benzoquinone, and phenyl-1, 4-benzoquinone, The phosphorus compounds which have the molecule interior division pole which comes to add a compound with pi bonds, such as a diazo phenylmethane and phenol resin Tetrapod phenyl phosphonium TETORAFENIRUBOREDO, triphenyl phosphine tetrapod phenyl BOREDO, etc. are mentioned. The object which added the quinone compound to organic phosphines or organic phosphines from a viewpoint of a moldability especially is desirable. The addition product of the third phosphine, such as triphenyl phosphine, a tree

p-tolyl phosphine, a tris (4-methylphenyl) phosphine, and a tris (4-methoxyphenyl) phosphine, and quinone compounds, such as 1, 4-benzoquinone, 1, and 4-naphthoquinone, is more desirable. Even if it uses independently, you may use these hardening accelerators combining two or more sorts.

[0025] Although the loadings of the Lynn system hardening accelerator are not restricted especially if they are an amount in which a hardening facilitatory effect is attained, its 0.005 - 5 % of the weight is desirable to a resin constituent, and they are 0.01 - 2 % of the weight more preferably. At less than 0.005 % of the weight, when there is an inclination to be inferior to hardenability and it exceeds 5 % of the weight, there is an inclination it to become difficult for a cure rate to be too quick and to obtain good mold goods.

[0026] Although powder mixing of them may be carried out after pulverizing the thermosetting resin, the phenol resin, and the hardening accelerator which have a dihydrobenzo oxazine ring by the mixer etc., they may carry out dissolution mixture in the method of carrying out melting mixture by the heating roller etc., and a solvent.

[0027] Moreover, to the above-mentioned constituent, a filler, strengthening fiber, a release agent, a coloring agent, adhesives, etc. can also be added if needed.

[0028] the resin constituent of this invention -- a heating roller etc. -- kneading -- after [ appropriate ] - 180-220 degrees C and fabrication -- thick -- the thermosetting resin which hardens compression molding or by carrying out transfer molding for 5 - 30 minutes by 2-7MPa, and has a dihydrobenzo oxazine ring -- independent -- it excels in hardenability more and a hardened material with still better mechanical characteristic and fire retardancy can be obtained

[0029] Moreover, the hardened material which has a better property is obtained by carrying out the postcure of this hardened material for 5 - 120 minutes at further 180-220 degrees C.

[0030] The thermosetting resin constituent of this invention may add an epoxy resin and its curing agent in uses, such as a metal tension laminate, a molding material, a sealing agent, and resin material for FRP, corresponding to the use.

[0031] As an epoxy resin used as an addition component There is especially no limit. A phenol novolak type epoxy resin, an orthochromatic cresol novolak type epoxy resin, A biphenyl type epoxy resin, bisphenol A, Bisphenol F The glycidyl ester type epoxy resin obtained by the reaction of polybasic acid and epichlorohydrin, such as Bisphenol S The glycidyl amine type epoxy resin obtained by the reaction of polyamine and epichlorohydrin, such as a diamino diphenylmethane the line oxidized and obtained by peroxy acids, such as a peracetic acid, in olefin combination -- there are an aliphatic epoxy resin and an alicycle group epoxy resin, a brominated epoxy resin, it is one-sort independent about these, or two or more sorts may be used together As a curing agent of an epoxy resin, although there is especially no limit, what does not generate volatile matter is desirable at the time of hardening, and polyaddition mold-curing agents, such as novolak type phenol resin, a dicyandiamide, imidazole derivatives, and triphenyl phosphine, are suitable for it to it especially. In addition, when using novolak type phenol resin as a curing agent of an epoxy resin, it is desirable that the hydroxyl of novolak type phenol resin adds the novolak type phenol resin of the amount which is usually 0.5-1.5 mols to one mol of epoxy groups of an epoxy resin in addition to the loadings of the novolak type phenol resin which indicated the above-mentioned thermosetting resin constituent.

[0032] The thermosetting resin constituent of this invention is suitably used as resin material, such as the highly efficient molding material which is excellent in thermal resistance, fire retardancy, fabrication-operation nature, etc., a paint, a coating material, adhesives, a sealing agent, a prepreg for printed circuit boards, a metal tension laminate, FRP, and a carbon product.

[0033] For example, a filler, strengthening fiber, a release agent, a coloring agent, etc. can be added remaining as it is or if needed, and the thermosetting resin constituent of this invention can be used as a sealing agent for closure of various molding materials, a semiconductor device, etc. Moreover, the prepreg used for manufacture of a printed circuit board can be obtained by infiltrating the varnish which contains a filler etc. if needed [ a thermosetting resin constituent and if needed ] for this invention into base materials, such as a glass fabric, and carrying out stoving. Let this prepreg be a metal tension laminate two or more sheet pile and by arranging metallic foils, such as copper foil, to the one side or both sides further, and carrying out heating pressurization. Thus, since it excels in thermal resistance and fire retardancy, the printed circuit board produced using the prepreg and metal tension laminate which are obtained is suitably used in various electrical and electric equipment and electronic fields.

[0034]

[Example] Next, although an example explains this invention, the range of this invention is not

limited to these examples.

[0035] The paraformaldehyde was suspended in the phenol, carrying out heating reflux using synthetic example 1 (composition of thermosetting resin (I) which has dihydrobenzo oxazine ring) bisphenol F 1.00kg (five mols), 95% paraformaldehyde 0.63kg (20 mols), and methanol 900ml. When suspension became 50 degrees C, aniline 0.93kg (ten mols) was divided, it added, reaction temperature was raised after the addition end, and the reflux reaction was performed. After reaction mixture emulsified, the reaction was continued for 2 hours, after the reaction end, a solvent and the subgenerated water were distilled off under reduced pressure, and the thermosetting resin which has a dihydrobenzo oxazine ring was obtained. The melt viscosity in 150 degrees C of the obtained thermosetting resin was 0.8P.

[0036] The paraformaldehyde was suspended in the phenol, carrying out heating reflux using synthetic example 2 (composition of thermosetting resin (II) which has dihydrobenzo oxazine ring) o-cresol 1.08kg (ten mols), 95% paraformaldehyde 0.63kg (20 mols), and methanol 900ml. When suspension became 50 degrees C, 4 and 4'-diamino diphenylmethane 0.99kg (five mols) was divided, it added, reaction temperature was raised after the addition end, and the reflux reaction was performed. After reaction mixture emulsified, the reaction was continued for 2 hours, after the reaction end, a solvent and the subgenerated water were distilled off under reduced pressure, and the thermosetting resin which has a dihydrobenzo oxazine ring was obtained. The melt viscosity in 150 degrees C of the obtained thermosetting resin was 1.1P.

[0037] After teaching synthetic example 3 (composition of novolak type phenol resin) phenol 2.4kg, 0.13kg [ of 37% formalin aqueous solution ], and paraformaldehyde 0.5kg, and 3g of oxalic acid to the 5L flask and making it react at reflux temperature for 4 hours, the interior was decompressed to 6666.1 or less Pa, an unreacted phenol and unreacted water were removed, and novolak type phenol resin was obtained. The melt viscosity in 150 degrees C of the obtained novolak type phenol resin was 2P.

[0038] the resin constituent obtained by [hardening] above -- grinding -- bore 120mmx80mmx4mm metal mold -- it was filled up inside, heating pressurization was carried out for 15 minutes by 180 degrees C and 1.96MPa, and the hardened material was produced

[0039] As the thermosetting resin which has examples 1-6, the example 1 of comparison - a 8 dihydrobenzo oxazine ring It blended by the composition which shows the Lynn system hardening accelerator (addition product of triphenyl phosphine and the 1, 4-benzoquinone), an imidazole, and a hexamethylenetetramine in Table 1 and Table 2, respectively as the thermosetting resin obtained in the above-mentioned synthetic examples 1 and 2, the novolak type phenol resin obtained in the above-mentioned synthetic example 3, and a hardening accelerator, the resin constituent of examples 1-6 and the examples 1-8 of comparison was produced, and the property was evaluated.

[0040]

[Table 1]

表1 配合組成

(単位: 質量部)

配合成分	実施例						比較例						
	1	2	3	4	5	6	1	2	3	4	5	6	
ジヒドロベンゾオキサジン樹脂(Ⅰ)	100	-	100	-	100	-	100	-	100	100	100	-	100
ジヒドロベンゾオキサジン樹脂(Ⅱ)	-	100	-	100	-	100	-	100	-	-	-	-	100
ノボラック型フェール樹脂	20	20	50	50	100	100	-	-	-	20	20	100	20
リン系硬化促進剤	2.4	2.4	3.0	3.0	4.0	4.0	2.0	2.0	-	-	-	-	-
イミダゾール	-	-	-	-	-	-	-	-	2.4	-	-	-	-
ヘキサメチレンテトラミン	-	-	-	-	-	-	-	-	2.0	-	2.4	2.0	-

Each following examination estimated the resin constituent of the produced example and the example of comparison, and its hardened material. An evaluation result is shown in Table 2.

- (1) It considered as time until it continues stirring in 1 time / 1 second and a cobwebbing is lost on the gel timer which warmed 0.3g of gelation-time resin constituents at 180 degrees C.
- (2) The three-point support type bending test based on JIS-K -6911 by the tensilon by Shimadzu Corp. was performed using the test piece fabricated on the conditions of 7MPa, and it asked for flexural strength (MPa), a bending elastic modulus (GPa), and \*\*\*\*\*-ed (%) for 180 degrees C of bending tests, and 90 minutes. Measurement was performed the condition for head speed 1.5mm/at a room temperature or 215 degrees C using the 80mmx10mmx3mm test piece.
- (3) It measured on condition that 5 degrees C of programming rates, and min using the test piece of a

5mmx3mmx3mm configuration by the apparatus for thermomechanical analysis (TMA-8141BS, TAS-100) by the glass transition temperature and coefficient-of-linear-expansion physical science electrical-and-electric-equipment incorporated company. It asked for the glass transition temperature (Tg, unit:degree C) from the folding point of a linear expansion curve, and asked for coefficient of linear expansion (E-5-/degree C) from the inclination below Tg.

(4) The resin board with a 50mm x50mm thickness [ of water absorptions ] of 3mm was produced, and it processed for 20 hours within PCT (pressure-cooker circuit tester) (121 degrees C, two atmospheric pressure), and asked from the weight change before and behind humidification.

[0041]

[Table 2]

【表 1】

項目	実施例						比較例							
	1	2	3	4	5	6	1	2	3	4	5	6		
ゲル化時間(秒)	25	25	20	20	15	15	60	60	80	25	25	30	140	140
室温 引張強度(MPa)	50	50	45	45	40	40	8.0	8.0	8.6	40	35	9.5	7.0	7.0
引張弾性率(GPa)	4.5	4.5	4.0	4.0	4.0	4.0	3.0	3.0	5.2	4.0	3.0	3.0	4.5	4.5
破断伸び率(%)	1.6	1.6	1.3	1.3	1.1	1.1	2.7	2.7	1.7	1.0	0.8	1.0	1.6	1.6
ガラス転移温度(℃)	128	128	128	135	135	138	138	155	120	132	150	145	146	
線膨張係数(E-5/℃)	6.3	6.3	6.5	6.5	5.3	6.3	6.6	7.4	9.0	7.0	6.6	6.5	6.6	
吸水率(%)	1.35	1.35	1.35	1.30	1.42	1.42	1.02	1.10	1.65	2.77	3.08	4.78	1.25	1.25

The gelation time is long and the examples 1-3 of comparison which do not contain the (B) component in this invention were inferior in hardenability. (C) The examples 4 and 5 of comparison using hardening accelerators other than a component had the high water absorption, although hardenability was good. Moreover, the example 6 of comparison which does not contain the (A) component also had the very high water absorption, although hardenability was good. (C) The gelation time is long and the examples 7 and 8 of comparison which do not contain a component were inferior in hardenability.

[0042] On the other hand, the examples 1-6 containing all (A) - (C) components of this invention had the short gelation time, the water absorption was also low, and excelling in a moldability and hardenability was shown.

[0043]

[Effect of the Invention] Since the hardened material which a moldability is good and is low hygroscopicity as the example showed the thermosetting resin constituent which becomes this invention, and its hardened material, and was excellent in hardenability and the mechanical characteristic is obtained, the industrial value is size.

[Translation done.]